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MONAZITE, THORIUM, AND CERIUM

BY

R. M. SANTMYERS

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By R. M. Santmyers[2]

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INTRODUCTION

Thorium and cerium are two of the group of elements known as "rare earth metals," and are obtained from the mineral monazite. Previous to the employment of thorium and cerium in incandescent gas mantles these elements were nothing more than laboratory curiosities, as the rest of the rare-earth metals are to-day. The most important salts of these rare earths are the nitrates. In the manufacture of gas mantles, thorium nitrate is the principal ingredient and forms from 98 to 99 per cent of the impregnating material; the other 1 or 2 per cent is cerium nitrate and is quite as necessary, for without it there would be little luminosity to the mantle when in use. When the mantle is "burned off," the nitrates are changed to oxides.

The monazite used as a source of thoria contains from 6 to 9 per cent of thorium oxide (ThO_2) and 24 to 32 per cent of cerium oxide (Ce_2O_3). As only the thoria and a very small part of the ceria from the monazite could be used in gas-mantle manufacture, for a time the residues consisting mostly of ceria with somewhat less lanthana and very much smaller quantities of erbia and other rare earths, found no commercial application. Later, however, they came to be utilized partly for the manufacture of cored flaming-arc carbons, a great improvement over the plain arc carbons used in electric lighting, and partly for the production of "cerium" metal, used in the manufacture of pyrophoric or sparking alloys or "flints" for automatic pocket lighters, miners' lamps, etc.

The almost universal use of electricity for lighting has resulted in a decided decrease in the manufacture of gas mantles, and therefore a decreasing supply of rare earth metals in the form of waste by-products is available.

Coincident with a decrease in the mantle industry there has been a large increase in the pyrophoric alloy industry in recent years and it seems probable that before long the quantity of rare earth metals obtained as a by-product of the gas-mantle industry will be insufficient for the manufacture of sparking alloys.

Thorium, like uranium, gradually breaks down and gives off helium, so that one of the proposed uses for monazite is as a source of helium gas. Helium has been obtained from monazite from Travancore, India, in the order of 1 c. c. per gram of monazite. It was thus estimated that for every 100 tons of monazite sand worked up, approximately 100,000 liters (1 liter = 61.022 cubic inches) of helium were allowed to escape into the atmosphere. It seems probable that these figures are high, for recent careful experiments on Japanese monazite of about the same ThO_2 content yielded only 0.246 c. c. per gram. To drive off the helium the monazite must be raised to a bright red heat. However, even in Europe where no suitable natural gas has yet been found, the oxtremely high cost of obtaining holium from monazite has made its production impracticable in view of the ease and cheapness of production of hydrogen which, though less desirable, is nevertheless suitable for filling all kinds of lighter-than-air craft.

Tungsten lamp filaments usually contain about 1 per cent of thoria (ThO_2). The thoria retards crystallization and thus prevents brittleness and greatly increases the luminosity of the filament.

In radio tubes the presence of thorium is actually necessary, since thorium is the material which emits electrons and causes the tubes to function. The "reactivation" or "rejuvenation" of radio tubes is simply the process of flashing them for a few seconds with a strong electric current, thus causing the surface of the filament to become coated with elemental thorium which is literally "boiled" to the surface from within the wire.

MINERALS AND ORES

Monazite.- The most important thorium-bearing mineral is monazite, which consists chiefly of the phosphates of cerium, lanthanum, and didymium (praseodymium and neodymium), including smaller and variable quantities of thorium, silicon, and other elements. Most monazites are yellow, but they vary in tint from a light greenish-yellow to dark honey-brown, and may be brownish-red or other colors. Monazite is semitransparent to opaque, and is characterized by a resinous or greasy luster. The specific gravity ranges from 4.9 to 5.3, and is generally over 5, so that it may be concentrated by simple gravity methods. The hardness (5 to 5.5) is about the same as that of steel and is considerably less than that of quartz. It is brittle and may show a distinct cleavage on breaking. In the original rock it may show sharp edges and plane crystal faces, but the commercial deposits are all in alluvial sand and the mineral is in rounded grains, intimately mixed with other heavy and resistant minerals, the most abundant of which are ilmenite, magnetite, garnet, and zircon.

The monazite sand as produced in this country generally carried less than 50 per cent monazite. Some process of concentration has been necessary to meet the requirements of the mantle industry, as usable monazite sand must carry a minimum of 6 per cent thoria, ThO_2. According to available sources of information, much of the crude sand produced in North Carolina carried less than 3 per cent of thoria, and analyses of sands contaning about two-thirds monazite showed the thoria content to range from 0.13 to 6.54 per cent. The average thoria content of 18 samples tested was only 2.60 per cent.[3]

Artificially concentrated monazite sand from the Centerville region, Idaho, yielded from 2.41 to 4.60 per cent of thoria, but some showed thorium oxide as high as 5.2 per cent.

Brazilian sand from the sea coast, as marketed, is said to run about 92 per cent monazite and to contain from 5 to 7 per cent thoria. The average content of thoria is about 6 per cent. The inland deposits are said to be slightly lower in their thoria content than the sea-coast sand, averaging from 4 to 5.7 per cent ThO_2.

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[3] Schaller, W. T., Thorium Minerals in 1916. U. S. Geol. Survey, Min. Res. of the United States, pt. 2, Sept. 13, 1917, p. 233.

The Indian sand found at Travancore carries as much as 46 per cent monazite, the remainder was ilmenite and zircon. A somewhat concentrated sand as marketed in England contained 62 per cent monazite, 26 per cent ilmenite (chiefly), and 11 per cent zircon (chiefly), and yielded from 5 to 6 per cent ThO_2. The Indian sand as marketed in the United States runs 90 per cent monazite and yields 9 per cent thoria.

The monazite sand from Ceylon is also rich in thoria. Sand averaging 48 per cent monazite yielded 4.15 per cent thoria, and 96 per cent monazite sand contained 8.39 per cent thoria. Artificial concentrations from pegmatites, with 45 to 87 per cent monazite, yielded from 5 to 7.3 per cent ThO_2.

The Malayan sands are unique in that they have a high cassiterite content, which ranges up to 65 per cent. The monazite in thses sands runs as high as 58 per cent.

Thorite.- The silicate of thorium, or thorite, is usually massive and irregularly disseminated through pegmatite in which it occurs in shapeless, compact masses. Rarely crystals are found similar in shape to those of zircon. Thorite is more or less opaque and has a dark brown or black color, a greasy luster, and a shell-like fracture. The rarely found clear yellow or orange crystals are known as orangite. The mineral is generally impure and contains considerable water. The formula of the mineral ($ThSiO_4$) calls for 81.5 per cent of thoria (ThO_2), but actual analyses have shown considerably lower percentages of thoria, ranging all the way from 49 to 74. The specific gravity of thorite is from 4.4 to 5.4, slightly under that of monazite.

Auerlite.- The mineral auerlite, named after Dr. Carl Auer von Welsbach, is dull yellowish-white to reddish in color and has a resinous luster. Like thorite, to which it is closely related, it is a silicate of thoria, but it also contains phosphorus in proportion represented by the formula $3ThO_2(3SiO_2.P_2O_5).6H_2O$. It is not certain, however, that the phosphoric acid belongs to the original mineral which, as found, is certainly more or less altered.

Thorianite.- The black mineral thorianite from the gem washings of certain districts of Ceylon is variable in composition, but ideally it has the formula ThO_2. Actually the content of thorium oxide reported in the analyses ranges from 59 to 93 per cent. It seems to be isomorphous with uraninite and contains uranium oxide in various quantities.

Thorianite forms small, simple cubes, usually only a few millimeters across and very rarely reaching an inch. Interpenetration twins, like those of fluorspar, are occasionally met with. The color is jet black, and when the surface is not water-worn and dulled, the luster is bright and pitchy. The streak is brown with a green tinge; thin sections are translucent and optically isotropic. The high specific gravity of about 9.5 is a striking feature, and its hardness is nearly 7.

Thorianite contains a higher percentage of thoria than any other known mineral (59 to 93 per cent).

Other thorium mineral.- A number of other minerals contain thoria in large percentages, but no large deposits of such minerals have been found. Thorogummite from Baringer Hill, near Llano, Tex., contains 41 per cent of thoria, ThO_2; that from Wodgina, Western Australia, 24 per cent. Mackintoshite from Texas carries 45 per cent of thoria, and that from Wodgina 25 per cent. Pilbarite from Wodgina has 31 per cent of thoria. A number of black or brown, heavy minerals with a greasy or pitchlike luster, such as fergusonite, zirkelite, uraninite, yttrocrasite, and yttrialite, contain various quantities of thoria.

Cerium minerals.- Cerite, a hydrous silicate of cerium metals, is the richest known cerium-bearing mineral. According to Rammelsberg's analysis made in 1859, it contain 64.55 per cent of Ce_2O_3, with 7.28 per cent of $(La,Di)_2O_3$ and approximated the formula, $2Ce_2O_3.3SiO_2.3H_2O$. The mineral is usually found as reddish granular masses much resembling in appearance certain varieties of corundum, from which, however, it is readily distinguished because its hardness (under 5½) is scarcely greater than that of ordinary pocket-knife steel. The specific gravity is 4.9. It was in this mineral that the element cerium was first discovered.

Allanite.- Allanite, sometimes called orthite, next to monazite is the most abundant cerium mineral. It is a brittle, black or black-brown, subtranslucent to opaque mineral, which contains the cerium metals, cerium, lanthanum, and didymium, and in smaller quantities the metals of the yttrium group. It has a hardness of 5.5 to 6, or a trifle more than that of cerite, but its specific gravity is much lower, 3.50 to 3.95. It has a small use as a source of cerium.

TESTS FOR IDENTIFICATION[4]

In connection with tests for thorium, it would be well to consider the other rare earth metals. The more important of these are, with their respective chemical symbols, lanthanum, La; praseodymium, Pr; neodymium, Nd; yttrium, Y; erbium, Er; and cerium, Ce. The group includes several other elements, but the definitive reactions for these rare substances are so obscure and difficult that no attempt will be made to give them under these tests.

To separate the rare earths they are precipitated together as hydroxides from acid solutions by means of ammonium or potassium hydroxides (this precipitation may often be omitted when it is known that calcium is absent). The precipitate when filtered and washed is dissolved in hydrochloric acid, the excess of acid removed by evaporation, the residue dissolved in water, and oxalic acid added; a precipitate of oxalates of the rare earths will then be thrown down, which is insoluble in oxalic acid. When filtered, washed and ignited, the precipitate yields oxides of the earths.

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[4] Brush, George J., and Penfield, S. L., Manual of Determinative Mineralogy New York, 1901, pp. 65-66. (With corrections made by R. C. Wells, U.S.G.S., Interior Department.)

In order to detect thorium the oxides are dissolved by heating with a few cubic centimeters of sulphuric acid; the solution is then evaporated until the excess of sulphuric acid is wholly driven off, thus converting the earths into normal sulphates. In order to regulate the heat it is best to place the crucible containing the sulphates inside a porcelain crucible, leaving an air space between, and to adjust the heat so that the outer crucible is not heated above a faint redness. The crucible should be covered toward the end of the operation, and the heating continued until no white fumes appear when the cover is raised. If the sulphates have been properly heated they should be wholly soluble in cold water, and thorium may then be precipitated from the dilute solution by adding sodium thiosulphate, $Na_2S_2O_3$, and boiling. The precipitate, when collected on a filter paper, washed, and ignited, yields thorium oxide, ThO_2. Zirconium, if present, will precipitate with thorium, but it should not be present after separation of the rare earths as oxalates. From solutions which are too concentrated, however, cerium may be precipitated with the thorium. To make certain, therefore, of the identity of the thorium, it will be best to convert the ignited material again into sulphate, and to repeat the precipitation with sodium thiosulphate.

In the cerium group, pure ignited oxide of cerium, CeO_2, is nearly white, as are also the oxides of lanthanum, La_2O_3, and didymium, Di_2O_3, but a mixture of cerium oxide with the latter always has a brown color. If the solution of the ignited oxides in sulphuric acid is yellow, it indicates cerium, and is due to ceric sulphate, $Ce(SO_4)_2$. After igniting the sulphates, however, cerous sulphate, $Ce_2(SO_4)_3$, is formed, which gives a colorless solution. If the oxides are dissolved in a borax bead in the oxidizing flame, a brownish-red or yellow bead, fading to yellow on cooling, indicates cerium. In the reducing flame the bead becomes colorless, or nearly so. With phosphorus salt the colors for cerium in the oxidizing flame are yellow when hot, fading to colorless when cold, and in the reducing flame, colorless both when hot and cold.

HISTORY

The mineral monazite was discovered in 1829 by A. Breithaupt, but it did not become of commercial significance until after the invention of the incandescent gas mantle. In 1885, an Austrian, Baron Carl Auer von Welsbach, while engaged upon research into the properties of rare earth minerals, discovered the brilliant light-emitting qualities of their oxides and soon conceived the basic idea of the incandescent gas mantle. The first successful mantles were made of oxides of lanthanum and zirconium, to which a little cerium oxide was added. Welsbach immediately secured patents in various countries and then sold the manufacturing rights.

About 1887 the industry took root in the United States with the formation of the Welsbach Light Co.

The early gas mantles proved somewhat of a failure. They consumed as much as 1 cubic foot of gas an hour and gave only about 10 candlepower. Welsbach was in danger of being discredited, and pushed his research further. It was found that thorium salts considerably improved the efficiency of the mantle, but as purer and still purer thorium salts were tried, a point was reached where no light whatever was emitted. Though the mantle should be impregnated mainly with thoria, the presence of 1 to 2 per cent of other oxides, chiefly cerium oxide, proved to be essential for best results.

Since thorium is only an accessory and a relatively minor constituent of monazite sand, quantities of ceria and other rare earth oxides remain as residues after extracting the thorium salts for the gas-mantle industry. It is the 6 to 10 per cent of thorium oxide (poorer sands are not used) for which the monazite sand is refined, and for many years the so-called "cerium oxides" or "mixed rare earth metal oxides" were considered as waste.

After a time it was found that these residues could be utilized for the production of misch metal (mixed metal), a sparking alloy which also goes by the name of "metallic cerium," although it really is a mixture of cerium, lanthanum, didymium, samarium and other closely allied metals of the rare earth group. It is not necessary to separate them, but the salts for making the metal must be purified. The process for making metallic cerium and the like is described in U. S. Patents 1,273,223, July 23, 1918, and 1,290,010, December 31, 1918, to Alcan and Marx Hirsch.

The development of the pyrophoric-alloy industry is likewise credited to Baron Auer von Welsbach, who succeeded in alloying the exceedingly soft cerium with iron in order to make it hard enough to give off sparks when scratched with a file or similarly roughened surface. He took out patents on this invention the world over, but in Germany and England the courts held that the patents did not cover nonferrous alloys, and were in fact restricted to iron alloys containing about 30 per cent of iron. The United States courts, on the other hand, have sustained the patents much more broadly to include any cerium-containing material hardened with any suitable alloying metal. When Welsbach and his associates developed the pyrophoric-alloy business, they did not form separate companies in each country, but centralized the manufacture of cerium and pyrophoric-alloys in the Treibacher Chemische Gesellschaft of Treibach, Austria.

It was not until after the outbreak of the war that cerium was made anywhere in the world outside the countries of the Central Powers, at which time an American company entered the field.[5]

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[5] Hirsch, Alcan, The American Pyrophoric-Alloy Industry. Chem. and Met. Eng., vol. 19, No. 6e, Sept. 28, 1918, pp. 510-512.

The coast sands were originally rich enough to be shipped as found, but these rich natural concentrations have been worked out and the sand is now artificially concentrated before being shipped.

India.-[6] Deposits of monazite sand especially rich in thoria were discovered in 1909 along the sea coast in the State of Travancore in the extreme southwestern part of India. Many of the soils and river sands show monazite, but only certain naturally concentrated deposits along the seacoast are of workable extent. Although there are numerous patches of beach carrying monazite sand in Travancore, there are only four which are considered of commercial size at present. The first deposit is on the east coast about 3 miles north of Cape Comorin. The sand is very fine, and the monazite content is sometimes as high as 20 per cent, though the average is much less, depending on wave action. Further southward and immediately north of the cape is the second deposit, much more extensive but of the same general character. The potential yield of these two deposits, together with the periodical accretion due to the action of the waves, is estimated at several thousand tons of commercial monazite.

The third deposit, and by far the richest, is on the west coast, immediately north of Cape Comorin. The extreme southern portion is very rich in monazite, the percentage rising to 50 or more. It has been estimated that 1,200 tons of monazite could be produced annually from this deposit for twenty years.

The fourth deposit is located on the west coast about 8 miles south of Trevandrum, the capital of the State, and is not quite as rich as the other three. Numerous other patches which may be of economic importance are interspersed in the regions between these four major fields.

Ceylon.- Immediately preceding the war, the Ceylon Government Mining Survey found substantial deposits of monazite sand near Bentota and near Kudremalai on the west coast of Ceylon. Samples of the Bentota sand contained as much as 47 per cent monazite and over 4 per cent thoria, ThO_2; by concentrating these electromagnetically to eliminate the ilmenite and zircon, a very high grade of monazite can be obtained containing nearly 10 per cent of thoria.

In addition to monazite, some thorianite has been produced in Ceylon, but only a very small amount has been sold. Thorianite and thorite are scarce compared with monazite, and even Ceylon's monazite deposits have been unable to compete with those of Travancore and Brazil.

Western Australia.-[7] Certain alluvial deposits in Western Australia contain monazite, cassiterite, and the rare earth minerals fergusonite, euxenite, and gadolinite. The monazite from Cooglegong is in small pebbles of an average weight of about one-fiftieth of an ounce. A sample of the sand from this district contained 80 per cent monazite and yielded 3.46 per cent thoria. The monazite sand from the residue obtained by resluicing low-grade alluvial tin ore contained 26 per cent monazite. The individual pebbles of monazite yielded a little more than 5 per cent of thoria.

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[6] Schaller, W. T., Thorium Minerals in 1916. U. S. Geol. Survey, Mineral Resources of the United States, pt. 2, 1916, pp. 228-229.

[7] Schaller, W. T., Work cited, p. 230.

Norway.-[8] The pegmatites of southern Norway contain large crystals and masses of monazite, which are saved by the feldspar miners and sold as a by-product. It is reported, however, that the quantity so obtained and sold has never been more than a ton for any one year.

Thorite, a thorium silicate mineral, occurs as small masses and indistinct crystals on the Langasund Fjord and near Arundal, in southern Norway.

Domestic Deposits

The workable deposits of monazite in the United States have been in North and South Carolina, Idaho, and Florida. The deposits in North Carolina were by far the most important, and have furnished the bulk of the domestic output.

North Carolina.- The commercial deposits of monazite found in North Carolina were along the eastern foothills of the Blue Ridge Mountains in the western part of the State, and extended into the northwestern part of South Carolina. The bulk of the supply of monazite was obtained from Rutherford, McDowell, Burke, Cleveland, and Lincoln Counties. The beds as worked are said to have averaged about 100 feet wide and from 300 feet to nearly a mile in length, and formed the sides and beds of numerous creeks in these counties. It was necessary in most cases to remove an overburden from 1 to 5 feet thick, some of which carried small amounts of monazite.

South Carolina.- In South Carolina the principal localities where monazite mining was carried on are in the vicinity of Cowpens, Spartanburg County, and near Gaffney, Cherokee County. The deposits were worked similarly to those in North Carolina. The known deposits in these two States are so small and scattered that no single deposit or group of deposits has been found which is large enough to justify the erection of a large-scale concentrating plant. The percentage of monazite in the gravel averages about one-fourth to 1 per cent. At the time the deposits were worked the mining was carried on by hand-sluicing methods similar to those employed for mining gold-bearing gravels, and usually by the farmers at odd times when there was no farm work to be done.

Idaho.- The deposits of monazite in the Centerville region, Boise County, are similar to those in the Carolinas. They are placer deposits and besides monazite contain ilmenite, garnet, and zircon. According to tests made, the sand contains from 0.1 to 0.15 per cent monazite, and the pure monazite contains only 5.2 per cent thoria. The material is gold-bearing and the monazite was caught as a by-product in the sluice-boxes, but the thoria content is too low to allow the monazite to be sold in competition with the Brazilian and Indian minerals.

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[8] Ibid, p. 230.

Florida.- The most recently operated monazite deposit in the United States was at Mineral City, 4 miles south of Jacksonville Beach, formerly known as Pablo Beach. This deposit was worked primarily for ilmenite, with zircon and rutile as by-products; the amount of monazite in the sand (a fraction of 1 per cent) was too small and the thorium content too low to work the deposits for monazite alone. During 1925 there was a reported production of 2,000 pounds. No monazite has been produced in Florida since that year, and it is now reported that the plant has been closed down.

MINING AND PREPARATION

As already noted, practically the entire output of monazite in the United States and foreign countries has been produced from monazite-bearing sands and gravels in which the monazite is found in varying percentages. In North Carolina unsuccessful attempts have been made to extract the mineral from the original rock matrix on a commercial scale.

The monazite sand deposits were worked somewhat like gold placer deposits, by sluicing and hydraulicking. In the Carolina monazite deposits sluicing was the method mostly used, and in many localities it has been the custom to sluice not only the gravels but all the overburden, inasmuch as even the top soil carries a small amount of monazite. In the stream gravels in favorable areas, the supply of monazite was replenished by the wash from the hillside soils during rains, especially where the hills have any considerable slope and the land was under cultivation. Under such conditions the stream gravels were worked by hand-sluicing two or more times a year.

Both wet and dry methods were employed for the separation of monazite from the sands and gravels. Sometimes a combination of the two methods was employed, the gravity concentrate produced in sluice boxes or by concentrating tables being dried and run through magnetic separators. The latter can be adjusted to clean the sand to 95 per cent monazite, though that shipped was not generally so carefully cleaned.

Hydraulicking has been introduced in the United States in a few localities where the gravels contain a quantity of gold, as in Idaho, but for the most part the mining of monazite has been done by hand methods, chiefly by farmers who in their spare time produced from a few pounds to a ton or more.

In Florida the heavy concentrates carrying monazite occur either on the surface of the beach or under a very shallow overburden of loose sand, so that the method of excavation was very simple. An attempt was made to mine the sand by dredging, but this proved unsuccessful, as sufficiently rich concentrates did not extend below water level.

The process for separating the minerals was very complicated, and involved the use of both the dry and wet methods, wet tables being used for the final separation of monazite.

Until recent years the mining and cleaning of monazite from the beach sands in Brazil and India has been exceedingly simple, as the action of the sea waves was sufficient to concentrate the monazite to a grade rich enough to be shipped in their natural condition. Eventually, however, it has been found necessary, especially in Brazil, to concentrate the sands by sluicing and electromagnetic separation methods similar to those employed in the United States.

Further treatment of monazite sand exported from both Brazil and India is necessary at the point of final consumption.

WORLD PRODUCTION

For a number of years prior to 1895 the United States furnished practically the world's requirements for monazite for the manufacture of incandescent gas mantles. The thorite deposits of Norway, however, were being worked to a limited extent, and these two countries were the only sources of thorium nitrate used by the German gas mantle industry prior to 1895. In 1895 Brazilian monazite suddenly appeared on the market in great quantities, and during the next two years the American monazite industry was almost at a standstill, producing only 15 tons and 32 tons in 1896 and 1897, respectively, as compared with 787 tons in 1895. In 1895 Norway also began to ship thorite in considerably larger quantities.

From 1898 on, the Brazilian monazite dominated the market, its output growing rapidly until in 1909 it reached the record figure of 7,121 tons. The industry in the United States, however, began to recover in 1898, and production again was on the upward trend, reaching 672 tons, or only 115 tons less than the all time record of 787 tons in 1895. In 1907, however, the production began to decline rapidly, and in 1911 there was no reported production of monazite sand in the United States. A few years later there was a slight revival in the industry, though the production never exceeded 19 tons in any year. This second decline was partly due to the production of monazite sand in Brazil and partly to the entrance of Indian monazite upon the world's markets.

Although the production of monazite in Brazil has been declining rapidly since 1911 and that of the United States has practically ceased, the production from British India has made rapid strides. This increase in the output of Travancore monazite was due to the relatively high monazite content of the sand and to the high thoria content of the concentrated product.

Even in India, however, production almost ceased in 1925, due to falling off in demand, and the revival during the next two years was made possible in large measure by a growing demand for ilmenite, which was also obtained from the monazite-bearing sands. Zircon is also produced from the same sands.

Ceylon began shipping monazite, thorianite, and thorite (the latter two in very small quantities) to England in 1918, but the production of monazite from that country has never exceeded 112 tons annually. No production has been reported since 1924.

The following Table 1 shows the world's production of monazite from 1893 to 1927.

Table 1. - World's production of monazite sand, 1893-1927[1]
(in short tons)

Year	United States	Brazil	India	Ceylon	Total	U. S. per cent of total
1893	65	- - -	- - -	- -	65	100
1894	273	- - -	- - -	- -	273	100
1895	787	3,307	- - -	- -	4,094	19
1896	15	192	- - -	- -	207	7
1897	22	294	- - -	- -	271	8
1898	125	2,149	- - -	- -	2,274	5
1899	175	2,932	- - -	- -	3,107	6
1900	454	1,633	- - -	- -	2,087	22
1901	374	1,811	- - -	- -	2,185	17
1902	401	1,328	- - -	- -	1,729	23
1903	431	3,636	- - -	- -	4,067	11
1904	372	5,357	- - -	- -	5,729	6
1905	672	4,891	- - -	- -	5,563	12
1906	423	4,797	- - -	- -	5,220	8
1907	274	4,891	- - -	- -	5,165	5
1908	211	5,473	- - -	- -	5,684	4
1909	271	7,121	- - -	- -	7,392	4
1910	50	5,994	- - -	- -	6,043	(2)
1911	- -	4,064	932	- -	4,996	- -
1912	- -	3,746	1,271	- -	5,018	- -
1913	- -	1,584	1,383	- -	2,967	- -
1914	- -	661	1,328	- -	1,989	- -
1915	18	484	1,241	- -	1,743	1
1916	19	- - -	1,448	- -	1,467	1
1917	11	1,252	2,173	- -	3,436	(2)
1918	- -	551	2,371	22	2,944	- -
1919	- -	161	2,267	- -	2,428	- -
1920	- -	1,270	1,838	81	3,189	- -
1921	- -	366	1,411	84	1,861	- -
1922	- -	127	140	112	379	- -
1923	- -	- - -	276	- -	276	- -
1924	- -	- - -	697	[3] 28	725	- -
1925	1	22	[4] .06	- -	23	(2)
1926	- -	221	72	- -	293	- -
1927	- -	224	314	- -	538	- -

1 Figures compiled from various official sources.
2 Less than 1 per cent.
3 Figures from the Imperial Institute, Statistical Summary, 1924-1926.
4 Figures from the Geol. Survey of India, vol. 59, pt. 3, 1926.

DOMESTIC PRODUCTION AND COSTS

The monazite industry of the United States has been mainly that of North and South Carolina, as these two States have accounted for practically all of the domestic output. Mining began in 1886, and the first shipment was made in 1887, when 12 tons were shipped from the Brindletown district of Burke County, North Carolina.

The Carolina deposits accounted for the entire output of monazite sand in the United States up to and including 1902. After that date Idaho became a small factor in the production of monazite sand, but even with this second producing area in operation, the output in the United States began to decline after 1905 until it practically ceased in 1911. The reason for this was partly depletion of the monazite deposits and partly competition from abroad. Imports in the form of monazite and thorium nitrate supplied domestic needs at lower cost. Added to the fact that foreign deposits were richer and larger, and that they could supply monazite more cheaply, was the further significant fact that the American monazite was leaner in thoria.

When the Great War broke out in 1914 and imports of monazite sand and thorium salts ceased, there was a slight revival in the monazite mining industry in the Carolinas, but this lasted only to 1917; since that time there has been no domestic production except for about 1 ton reported in 1925 from the deposit 4 miles south of Pablo Beach (Jacksonville Beach) near Jacksonville, Fla.

The following Table 2 shows the production of monazite sand in the United States from 1893 on.

Table 2. - Production of monazite sand in the United States, 1893-1928
(in short tons)[1]

Year	North Carolina		Other States		Total	
	Quantity	Value	Quantity	Value	Quantity	Value
1893	65	$ 7,600	- -	- - -	65	$ 7,600
1894	273	36,193	- -	- - -	273	36,193
1895	787	137,150	- -	- - -	787	137,150
1896	15	1,500	- -	- - -	15	1,500
1897	22	1,980	- -	- - -	22	1,980
1898	125	13,542	- -	- - -	125	13,542
1899	175	20,000	- -	- - -	175	20,000
1900	454	48,805	- -	- - -	454	48,805
1901	374	59,262	- -	- - -	374	59,262
1902	401	64,160	- -	- - -	401	64,160
1903	387	58,694	44	$5,936	431	64,630
1904	343	79,438	29	5,400	372	64,838
1905	2/ 447	107,324	225	54,984	672	162,308
1906	3/ 349	125,510	74	26,802	423	152,312
1907	3/4/ 228	54,824	46	10,930	274	65,754
1908	3/4/ 155	37,224	56	13,494	211	50,718
1909	3/4/ 196	46,928	75	18,104	271	65,032
1910	42	10,104	8	1,902	50	12,006
1911	- -	- - -	- -	- - -	- -	- - -
1912	- -	- - -	- -	- - -	- -	- - -
1913	- -	- - -	- -	- - -	- -	- - -
1914	- -	- - -	- -	- - -	- -	- - -
1915	18	3,600	- -	- - -	18	3,600
1916	19	3,400	- -	- - -	19	3,400
1917	- -	- - -	11	1,760	11	1,760
1918	- -	- - -	- -	- - -	- -	- - -
1919	- -	- - -	- -	- - -	- -	- - -
1920	- -	- - -	- -	- - -	- -	- - -
1921	- -	- - -	- -	- - -	- -	- - -
1922	- -	- - -	- -	- - -	- -	- - -
1923	- -	- - -	- -	- - -	- -	- - -
1924	- -	- - -	- -	- - -	- -	- - -
1925	(5)	- - -	1	120	1	120
1926	- -	- - -	- -	- - -	- -	- - -
1927	- -	- - -	- -	- - -	- -	- - -
1928	- -	- - -	- -	- - -	- -	- - -

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1 Mineral Resources of the United States and U. S. Geol. Survey publications.

2 1905 and 1906, Mineral Industry of North Carolina.

3 In 1906 South Carolina produced 74 short tons of monazite sand, valued at $26,802; 1907, 46 short tons, valued at $10,930; 1908, 56 short tons, valued at $13,494; 1909, 75 short tons, valued at $18,104.

4 Figures for 1907-1909, inclusive, were collected jointly by the U. S. Geol. Survey and North Carolina Geological Econ. Survey.

5 In 1925 production was for Florida alone (value estimated).

During the 10-year period from 1900 to 1909, when the domestic industry was at its height, the cost of producing monazite in the United States was considerably higher than that for the cost of importation from Brazil. The cost of producing 1 ton of domestic concentrates containing 92 to 95 per cent monazite was estimated to be approximately $169 at the concentrating plant, whereas during this period Brazilian monazite was laid down at the German ports at prices ranging from $95 to $120 per ton (5 per cent thoria), or a difference of from $45 to $70 per short ton. When the Indian monazite first came on the market in 1911, American monazite was costing approximately $260 per ton, and Brazilian monazite was being sold on the world markets at approximately $133 per ton. The Indian monazite, however, was exported at a price ranging from $120 to $126, or about one-half the price of the American product, and it carried over 50 per cent more thoria (ThO_2) than the American monazite.

Although there was still monazite sand in the United States, it could not be produced in competition with foreign monazite, and the industry in this country was forced to close down. Even the Brazilian monazite, which is richer in thoria than the American monazite, found competition with the still richer Travancore monazite very difficult.

IMPORTS

Before 1909 imports of monazite into the United States were negligible in amount, but in that year monazite began to be imported in increasing quantities. During 1909, 35 short tons, valued at $8,324 or $238 per ton, were imported, but in 1910 there was an increase to 227 tons valued at $39,699 ($175 a ton), and during the next year it increased further to 353 tons, valued at $60,542 ($172 a ton).

The duty of 4 cents a pound under the Payne-Aldrich Tariff Act of 1909, was equivalent, roughly, to 50 per cent of the foreign value, hence the 25 per cent rate provided in the Underwood Act of 1913 represented a further substantial reduction. Due to the virtual absence of domestic production and expanding consumption, imports increased rapidly, amounting in 1927 to 2,914 short tons, or more than seven times the quantity imported in 1913 and more than four times the maximum production (1905) of the United States.

As a result of the decline in the production of gas mantles since 1918, the imports of monazite have fallen off considerably, as may be seen from the following Table 3.

Under the Tariff Act of 1922, monazite sand has been on the free list, but the duties on thorium nitrate and other thorium products were increased. This has resulted in an increased importation of monazite sand, and a decrease in the importation of thorium nitrate.

With the increase in the imports of monazite sand, the United States became an important factor in the production of thorium salts. The manufacture of thorium and cerium nitrate expanded rapidly until about 1920, when our requirements for these chemicals were almost wholly furnished by imports. With monazite placed on the free list under the Tariff Act of 1922, the thorium and cerium nitrate industry again flourished in this country and imports fell off.

Table 3. - Monazite sand imported for consumption in the United States, 1901-1928.[1]

Year	Quantity (short tons)	Value
1901	- - -	- - -
1902	[2] .1	$ 12
1903	- - -	- - -
1904	- - -	- - -
1905	- - -	- - -
1906	- - -	- - -
1907	- - -	- - -
1908	- - -	- - -
1909	35	8,324
1910	227	39,699
1911	353	60,542
1912	278	47,334
1913	409	65,848
1914	385	61,595
1915	937	161,456
1916	1,218	188,383
1917	2,914	355,017
1918	1,497	204,661
1919	316	48,768
1920	907	141,528
1921	.03	40
1922	1,733	210,187
1923	127	11,334
1924	273	18,320
1925	- - -	- - -
1926	335	23,816
1927	597	41,029
1928	442	24,151

[1] Mineral Resources of the United States.

[2] Includes thorite.

TARIFF HISTORY

The following Table 4 shows the tariff classifications for monazite sand, thorium and cerium minerals and metals, incandescent mantles and mantle scrap, and thorium and cerium nitrate and other rare metal salts and compounds.

Table 4. - Tariff classifications

Act of-	Paragraph	Tariff classification	Rates of duty specified and ad valorem
1890	530	Cerium	Free
1894	60	Concentrated thorium nitrate solution was held dutiable as a chemical compound or salt	25% ad valorem
1894	432	Cerium	Free
1897	183	Monazite sand and thorite	6¢ per pound
1897	518	Cerium	Free
1909	183	Monazite sand and thorite	4¢ per pound
		Thorium, oxide of and salts of	
		Gas mantles treated with chemicals or metallic oxides, and gas mantle scrap consisting in chief value of metallic oxide	40% ad valorem
1909	530	Cerium, cerite, or cerium ore	Free
1913	154	Monazite sand and thorite; thorium, oxide of and salts of.	
		Gas, kerosene, or alcohol. Mantles treated with chemicals or metallic oxides	25% ad valorem
		Gas mantle scrap consisting in chief value of metallic oxides	10% ad valorem
1913	445	Cerium, cerite, or cerium ore	Free
1922	89	Thorium nitrate, thorium oxide, and other salts of thorium not specially provided for, cerium nitrate, cerium fluoride, and other salts of cerium not specially provided for, and gas-mantle scrap consisting in chief value of metallic oxides	35% ad valorem
1922	302	Cerium metal	$2 per pound
		Ferrocerium and all other cerium alloys	$2 per pound and 25% ad valorem
1922	1544	Cerite or cerium ore	Free
1922	1621	Monazite sand and other thorium ores	Free

EXPORTS

The United States, before the discovery of the richer Brazilian deposits, was the leading world factor in the production of monazite, supplying not only our own markets but substantially all that was consumed in the thorium nitrate industry in Europe. While no figures are recorded, it is understood that from one-fourth to one-third of the domestic production of monazite sand was annually exported to England and Germany and there used for the manufacture of thorium salts, principally nitrate. Of course, an export trade could not be maintained against the plentiful supply of cheaper and richer monazite which began to come from Brazil in 1895 and which eventually stifled even the home trade in domestic monazite.

MONAZITE TRADE IN FOREIGN COUNTRIES

Brazil.- The mining of monazite sand was started in Brazil by an American, John Gordon, who realized a considerable profit by shipping the product to Germany as ballast. In 1903 the Brazilian government decided that the deposits of this mineral along the sea coast belonged to the State, and forbade their free exploitation. Bids were let and the A. C. de Freytas Co., of Hamburg, secured the rights to exploit these deposits, but later the company formed a partnership with John Gordon, the original shipper of this mineral. They then sold their entire output to the German thorium syndicate, and in this manner held a partial monopoly over the production of monazite in Brazil.

It was soon found out that other manufacturers of thorium products were able to obtain supplies elsewhere, and that the whole output from Brazil could not be controlled; so the German syndicate decided to eliminate all competition. Accordingly, in January, 1906, the price of thorium nitrate was reduced by nearly one-half. Sales were made only to consumers, and each consumer was permitted to buy only such quantities as he could reasonably be expected to require for his own needs, leaving no surplus for resale.

The result of this cut in price was to eliminate the smaller manufacturers of thorium nitrate. Although it had little effect upon the larger well-established firms mining their own monazite, the formation of this cartel and the reduction in the price of thorium nitrate had considerable bearing on the export trade of monazite from Brazil. Prior to 1903 the exports never exceeded 3,400 short tons annually, but after that date the export trade in this mineral, as may be seen in Table 5, increased rapidly until in 1909, it reached its peak, 7,121 tons, valued at $704,387. By 1912 the exports of monazite sand had fallen to about 3,700 tons, and after that date they never exceeded 1,600 tons annually.

During the period when the exports of monazite sand had reached their peak, it was estimated that Germany took about two-thirds of the total, the balance going to France, United States, and England, in amounts decreasing in the order named.

Table 5. - Monazite sand exported from Brazil, 1895-1927[1]

Year	Quantity (short tons)	Value	Average value per ton
1895	3,307	- - -	- -
1896	192	- - -	- -
1897	249	$ 21,602	$ 87
1898	2,149	92,775	43
1899	2,932	- - -	- -
1900	1,633	165,412	101
1901	1,811	165,154	91
1902	1,328	261,141	197
1903	3,636	360,797	99
1904	5,357	529,597	99
1905	4,891	486,435	99
1906	4,797	478,961	100
1907	4,891	486,066	99
1908	5,473	551,744	101
1909	7,121	704,387	99
1910	5,994	620,605	104
1911	4,064	540,688	133
1912	3,746	528,614	141
1913	1,584	186,520	118
1914	661	79,068	120
1915	484	56,358	117
1916	- - -	- - -	- -
1917	1,252	- - -	- -
1918	550	- - -	- -
1919	161	- - -	- -
1920	1,270	- - -	- -
1921	366	- - -	- -
1922	127	- - -	- -
1923	- - -	- - -	- -
1924	- - -	- - -	- -
1925	22	- - -	- -
1926	221	- - -	- -
1927	224	- - -	- -

[1] Mineral Resources of the United States, 1895-1916; Bull. Imperial Inst., 1917-1922; Imperial Inst. Statistical Summary, 1923-1927.

India.- Monazite from Travancore, India, made its first appearance on the world markets in 1911. The officially reported export figures of monazite sand from India for that year were 932 short tons, valued at $117,010. Although the exports of this mineral from India never exceeded 2,400 tons in any one year they came on the market after the peak had been passed, for electric lighting was already superseding gas, gasoline, and kerosene lighting, and the mantle industry was beginning to decline - a decline which is still progressing. The Travancore production of monazite therefore seriously diminished the export trade of Brazil, whose shipments had already begun to drop off sharply in 1910.

Prior to the war the output, which was obtained from beach sands between Muttum and Colachel, in Travancore, was in the hands of a firm which operated under German control. After the outbreak of the war this German interest was soon eliminated, and the industry was placed under the control of British capital.

In recent years the trade in monazite sand from India has followed the general downward trend of other producing countries.

Owing to lack of shipping facilities in Travancore, the sand at present is not shipped direct, but is transported to Tuticorin, on the extreme southeastern tip of the peninsula, in small coasting vessels, except during the monsoon, when it is carried overland to Quilon on the eastern coast. It is exported from Tuticorin in bags of 112 pounds each.

Table 6 shows the export trade in monazite sand from Travancore, India, for the years 1911 to 1927.

Ceylon.- In 1918 the Ceylon Government purchased a magnetic separator for the purpose of concentrating the sand which was a waste product from gem washing, and during that year shipped about 22 tons to England. Shipments from that date until 1922 increased slightly, but ceased entirely after 1924.

Table 6. - Monazite sand exported from Travancore, India, 1911-1927[1]

Year	Quantity (short tons)	Value	Average value per ton
1911	932	$117,010	$126
1912	1,271	301,566	159
1913	1,383	204,451	148
1914	1,328	201,527	152
1915	1,241	161,753	130
1916	1,448	183,505	127
1917	2,173	274,904	127
1918	2,371	286,243	121
1919	- - -	- - -	- -
1920	1,053	- - -	- -
1921	- - -	- - -	- -
1922	22	- - -	- -
1923	230	- - -	- -
1924	112	- - -	- -
1925	319	- - -	- -
1926	388	- - -	- -
1927	563	- - -	- -

[1] Mineral Resources of the United States, 1911-1919; Imperial Inst. Statistical Summary, 1920-1927.

MARKETS AND PRICES

The demand for monazite sand in the United States is limited to the incandescent gas mantle and the pyrophoric-alloy industries. Requirements for the manufacture of gas mantles have declined rapidly in recent years, while those for the manufacture of "flints" or sparking metal have increased considerably due to the increased use of automatic cigar lighters, gas lighters, miners' lamps, etc.

Despite the attempted monopoly of the manufacture of thorium nitrate by German and Austrian firms, France and the United States for some time previous to the outbreak of the World War had been manufacturing this product chiefly from Brazilian and American monazite. As a result of the war and the serious shortage of thorium nitrate caused by the stoppage of shipments from Germany, other countries took up the manufacture of this product, requiring exceptionally large quantities of monazite sand.

The price of monazite sand has fluctuated considerably since the war, in most cases reflecting the declining use of this mineral. During the war the price of monazite sand delivered to New York, exclusive of the duty of $80 a ton, varied from $25 to $40 per unit (1 per cent per short ton) of thoria, or from $150 to $240 per ton. Immediately after the war the price was quoted (November, 1919) at $27 per unit, or about $162 per ton of the 6 per cent (thoria) sand, duty not paid.

Before the end of 1925 the price had dropped to $120 per ton (6 per cent ThO_2). After increasing slightly during the latter part of 1928 to $130 per ton, the price fell to $60 a ton during the late fall of 1929, the lowest figure so far recorded in the history of the industry.

Table 7 following shows the prices of monazite sand (per ton, minimum 6 per cent ThO_2) at New York for the years 1925-1929.

Table 7. - Prices of monazite, January, 1920-November, 1929[1]
(Per ton, minimum 6 per cent thoria)

Month	1920	1921	1922	1923	1924	1925	1926	1927	1928	1929
Jan.	-	$180	$ 180	$120-160	$120-160	$120-160	$120	$120	$120	$130
Feb.	-	180	180	120-160	120-160	120-160	120	120	120	130
March	$228-252	180	180	120-160	120-160	120-160	120	120	130	130
April	252	180	180	120-160	120-160	120	120	120	130	130
May	252	180	180	120-160	120-160	120	120	120	130	80
June	252	180	160-200	120-160	120-160	120	120	120	130	80
July	252	180	120-160	120-160	120-160	120	120	120	130	80
Aug.	210	180	120-160	120-160	120-160	120	120	120	130	80
Sept.	210	180	120-160	120-160	120-160	120	120	120	130	80
Oct.	210	180	120-160	120-160	120-160	120	120	120	130	60
Nov.	210	180	120-160	120-160	120-160	120	120	120	130	60
Dec.	180	180	120-160	120-160	120-160	120	120	120	130	60

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[1] Engineering and Mining Journal.

INCANDESCENT GAS MANTLES

Although the incandescent gas mantle was first patented in 1884, it was not until about 1893, when it was discovered that the maximum luminosity was produced by employing a mixture composed of 98 to 99 per cent thoria and 1 to 2 per cent of ceria, that the incandescent gas mantle came into extensive commercial use, and it was not until a few years later that sufficiently large deposits of the raw material (monazite) were found to assure the commercial success of this mantle. A rapid spread of lighting by incandescent gas burners followed, and later the use of these mantles undoubtedly helped to sustain the gas companies through the critical period during which electricity was replacing gas in the lighting of homes, offices, factories, and streets.

About 1900 the inverted mantle was invented. This form throws the light downward instead of upward, and avoids a shadow, so that it was an important advanc in the industry.

Method of Manufacture

The gas mantle is generally woven of cotton, ramie, or artificial silk fiber into the form of a hose or tube, bleached, and very thoroughly washed in distilled water in order to decrease the quantity of mineral matter and to remove grease and other foreign substances.[9]

After drying in a current of hot air the fabric is then treated with the "rare earth" nitrates, which is a solution containing from 25 to 50 per cent of thorium and cerium nitrate in the proportion of about 98 to 99 parts of thorium nitrate to 1 to 2 parts of cerium nitrate. Small quantities of beryllium nitrate and magnesium nitrate are also added to the solution to strengthen the ash skeleton. The fabric is then cut into suitable lengths and one end closed and sewed with asbestos thread. After this the mantle is "fixed", a process which strengthens the parts of the mantle which are subjected to the greatest strain. This is accomplished by treating these portions with a thorium solution containing a considerable proportion of alumina and alkaline earths. If desired, the mantle may then be branded with the manufacturers' trade-mark by the use of a mixture containing didymium nitrate, which is a by-product of the manufacture of cerium nitrate. After the fabric is dried and shaped, it is "burned off", and the nitrates are converted into the oxides by an intense flame. This operation requires considerabl skill and 2 or 3 per cent of the total number of upright mantles are usually damaged in the process. In the case of the inverted mantle the loss may run as high as 10 to 20 per cent. This gives rise to the material known as gas mantle scrap which is mostly obtained in this manner by the manufacturers; but the additional scrap is obtained in the form of worn-out mantles collected from the gas companies and other large consumers. The mantles which are not damaged in the heating process and have passed inspection for size, shape, and material defects, are dipped into a solution of collodion, in order to give them the necessary strength to withstand the shocks of shipping.

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[9] U. S. Tariff Commission, Incandescent Gas Mantle Industry. Tariff Information Survey, Para. 154, Tariff Act 1913, - C22, 1921.

The rare earth nitrates may be replaced in the fixing process by the corresponding acetates or formates, and if of artificial silk the impregnated stocking is treated with ammonia or some volatile alkali such as hydrazine or tetraethyl ammonium hydroxide in order to produce within the fibers the hydroxides of thorium and cerium. Hydrogen peroxide may also be employed as the precipitant.

From a kilogram of thorium nitrate 500 to 600 mantles can be made, hence each mantle contains about 0.5 grams of thoria.[10]

Although in most instances the optimum effect is attained by mixing the thorium and cerium salts in such proportions that the mixed oxides of the mantle consist of 98.8 per cent of thoria and 1.2 per cent of ceria, yet owing to the yellow color of the light produced by this amount of ceria in inverted mantles, it is customary in this form of illumination to reduce the proportion of ceria to from 0.5 to 0.7 per cent.

Mixtures of ceria and thoria have a catalytic action on the combination of hydrogen and oxygen which reaches a maximum at 450° C. when the mixtures contain 1 per cent of ceria. This was the mixture finally arrived at by Welsbach to secure the highest illuminating power. The activity, or in other words, the illuminating power, of pure thoria is the same as that of a mixture containing 9 per cent ceria.

It has been considered that the Welsbach mixture represents the best illuminant obtainable from the two oxides, thoria, and ceria.

The Gas-Mantle Industry

There are no recent figures available concerning the production or consumption of incandescent gas mantles in the United States. It has been estimated, however, that immediately before the war the United States had an annual consumption of approximately 60 million mantles, which after the war had increased to about 80 million mantles. From available figures concerning the importation of monazite sand and thorium nitrate it would seem that the consumption of incandescent gas mantles has decreased approximately 50 per cent from the above estimate.

With the important improvements in the electric lamp, the tremendous network of hydroelectric and steam-generated electric power lines throughout the country, and the manufacture of small gasoline-driven lighting plants for isolated places, there has been a great falling off in the use of incandescent gas mantles.

The following is a list of producers of incandescent gas mantles in the United States.

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[10] Thorp, Edward, A Dictionary of Applied Chemistry, vol. 7, London, 1927, p. 40.

MANUFACTURERS OF INCANDESCENT GAS MANTLES

Lindsay Light Co.,
161 E. Grand Ave., Chicago, Ill.

National Stamping and Electric Works,
3220 West Lake St., Chicago, Ill.

Sunshine Mantle Co.,
Chanute, Kans.

American Gas Machine Co.,
Albert Lea, Minn.

Hickory Gas Mantle Manufacturing Co.,
University and Driggs St., St. Paul, Minn.

General Gas Mantle Co.,
Camden, N. J.

Welsbach Co.,
200 Hudson St., New York, N. Y.
Gloucester, N. J.

Arrow Light Co.,
773 Cony Island Ave., Brooklyn, N. Y.

Buffalo Gas Mantle Co.,
407 Master St., Buffalo, N. Y.

Louis Deutsch,
177½ E. 87th St., New York, N. Y.

Rapid Manufacturing Co.,
799 Broadway, New York, N. Y.

E. J. S. Van Houten,
290 Broadway, New York, N. Y.

Incandescent Light & Stove Co.,
Madison & Ebersole Ave., Cincinnati, Ohio.

Harris Wood Fiber Mantle Co.,
3805 Park Ave., Cleveland, Ohio.

Erie Gas Mantle Manufacturing Co.,
Erie, Pa.

R. E. Tongue & Bros. Co.,
Allegheny Ave. & Amber St., Philadelphia, Pa.

I. Sniderman Co.,
1705 Center Ave., Pittsburgh, Pa.

Mantle Lamp Co. of America,
Chicago, Ill.

Imports and Exports

Tables 8 and 9 show the foreign trade of the United States in incandescent mantles (gas, gasoline, kerosene, etc.) for the years 1917 to 1927.

Table 8. - Incandescent mantles imported for consumption in the United States, 1918-1928[1]

Year[2]	Number	Value
1918	3,117	$ 316
1919	132	54
1920	208,497	13,243
1921	621,968	27,001
1922	2,407,911	92,713
1923	2,067,137	65,816
1924	2,061,000	54,507
1925	2,584,848	60,980
1926	1,664,976	43,408
1927	1,066,080	24,947
1928	- - -	- - -

[1] Commerce and Navigation of the United States, Bureau of For. and Dom. Com., Dept. of Commerce.

[2] Classified as mantles, gas, kerosene, or alcohol, treated with chemicals or metallic oxides, 1918-1922, then classified as incandescent mantles, treated with chemicals or metallic oxides, 1923-1925.

Table 9. - Domestic exports of incandescent mantles, 1917-1928[1]

Year	Value					
	Canada	British India	Hong Kong	Australia	All other	Total
1917	- - -	- - -	- - -	- - -	- - -	[2]/$122,660
1918	$90,246	$22,043	$7,874	$10,726	$121,395	252,284
1919	70,654	31,073	6,817	23,308	226,790	358,642
1920	78,359	74,404	16,342	58,125	251,141	478,371
1921	89,599	46,791	2,394	7,538	57,942	204,264
1922	53,155	50,787	925	10,917	49,160	164,944
1923	42,313	48,277	6,610	10,534	39,102	146,836
1924	39,805	47,814	6,603	11,818	48,264	154,304
1925	35,283	42,946	6,486	18,939	52,239	155,893
1926	30,401	40,275	5,839	30,149	73,114	179,778
1927	28,904	46,016	8,405	8,527	53,324	145,176
1928	- - -	- - -	- - -	- - -	- - -	202,275

[1] Commerce and Navigation of the United States, Bureau of For. and Dom. Com., Dept. of Commerce.

[2] Figures cover period beginning July 1.

GAS-MANTLE SCRAP

In the manufacture of incandescent gas mantles the converting of the thorium and cerium nitrates into oxides by a process known as "burning off" requires considerable skill. From 2 to 3 per cent of the upright mantles and from 10 to 20 per cent of the inverted mantles are damaged in the process. These mantles, together with worn-out mantles collected from gas companies and other large consumers, carry considerable thorium and cerium salts. They are reduced to ashes by a firing process, and the ashes are further treated for the recovery of the valuable thorium and cerium oxides.

Imports

Table 10 shows the imports for consumption into the United States of gas-mantle scrap for the years 1910 to 1922.

Table 10. - Imports for consumption of gas-mantle scrap, 1910-1922[1]

Year	Quantity (pounds)	Value
1910 [2]	- - -	$2,892
1911	- - -	6,120
1912	- - -	2,631
1913	- - -	2,198
1914	60,435	3,525
1915	4,175	228
1916	3,600	190
1917	- - -	- - -
1918 [3]	44	111
1919	- - -	- - -
1920	144	364
1921	- - -	- - -
1922	590	245

[1] Commerce and Navigation of the United States, Bureau of For. and Dom. Com., Dept. of Commerce.

[2] Fiscal years, 1910-1916; calendar years, 1917-1922.

[3] Mantle scrap consisting chiefly of metallic oxides.

PROPERTIES OF THORIUM

Thorium as usually prepared is a grayish metallic powder consisting of small, thin, hexagonal, crystallized leaflets. The larger crystals are said to be as white and lustrous as silver; the smaller ones are gray and less lustrous. They are somewhat brittle.

Thorium is unchanged in air up to 120° C., but at higher temperatures it burns with an intense white light to thorium oxide, ThO_2. The metal is fusible at 1845° C., somewhat ductile and malleable, and when finely powdered it ignites in air under slight friction. Thorium is strongly radioactive; glows and forms a salt when heated in chlorine, bromine, iodine, or sulphur; the solution of its salts are colorless. It has a specific gravity of 11.2 and a boiling point above 3,000° C.

THORIUM NITRATE

Description and Uses

Thorium nitrate is a white crystalline material, very soluble in water and alcohol. For use in gas mantle manufacture it must be thoroughly purified; the presence of phosphates in particular causes the mantle to be excessively brittle and not more than 0.004 per cent P_2O_5 is usually permitted. The presence of alkalies and alkaline earths causes the mantles to shrink and lose their shape, and the presence of certain rare earth metals decreases their luminosity.

As already stated, by far the most important use for thorium nitrate is in the manufacture of incandescent gas mantles; thorium nitrate with about 1 per cent cerium nitrate constitutes the impregnating fluid used for all types of mantles.

A small amount of thorium metal powder is sometimes alloyed with tungsten to be used as a filament for incandescent electric lights, and certain flash-light powders also contain thorium. Small pastilles consisting of 99 per cent of thoria and 1 per cent of ceria give off an intense white light when heated in a gas flame, and are used for motion picture and other projection lanterns in localities where the electric arc is not available. They are similar in many respects to the so-called "calcium" lights formerly well known.

Method of Manufacture

The preparation of pure thorium nitrate is a long and complex process, and there is no recognized procedure used by all manufacturers. The general principles involved are well known, but certain of the details of the final stages in the purification are guarded as trade secrets.

The first step in the process is known as "breaking the monazite sand," and consists of heating the finely ground sand with slightly more than its weight of sulphuric acid. This changes the phosphates to sulphates and yields a white pasty mass which is poured into water and allowed to settle. The insoluble residue is filtered off, and the solution, containing the thorium, cerium, lanthanum, etc., in dilute phosphoric and sulphuric acids, is partly neutralized with an alkali or alkaline earth. This causes the precipitation of a much enriched thorium phosphate, and by filtering, a large proportion of the other rare earths is removed. This filtrate contains the cerium, which is separated from the salts of other similar elements by another series of operations. The thorium phosphate precipitate is usually redissolved in a small quantity of acid, and the process is repeated. In the older practice the partly purified phosphate was next treated with oxalic acid and the thorium precipitated as thorium oxalate. In the more recent processes, however, the phosphate is cooked with soda ash to convert it to an insoluble basic carbonate which when treated with caustic soda yields thorium hydroxide. The hydroxide is dissolved in sulphuric acid and the thorium sulphate fractionally crystallized, a process which removes most of the more soluble cerium sulphate. The sulphate is reconverted to the hydroxide with ammonia and the thorium nitrate finally obtained by dissolving the ammonia precipitate in pure nitric acid and crystallizing. The process is said to yield at least 90 per cent of the thorium contained in the monazite sand.

Imports and Exports

The imports of thorium nitrate into the United States did not assume major proportions until the formation of the German-Austrian Thorium Cartel in 1902. In that year the Brazilian Government sold the rights to exploit the beach sands to a German syndicate, which during the following year formed a partnership with the original producer, an American. The French, however, were already producing monazite sand in Brazil, exploiting the deposits which lay behind those held by the Brazilian Government. This cartel rigidly enforced the policy of price-cutting with regard to thorium nitrate and the elimination of producers of this compound outside of the cartel. This policy was reflected in the increased importations of thorium nitrate into the United States, which tended to depress the domestic mining industry.

By 1909 the imports had reached the high figure of 127,833 pounds, valued at $236,057. With the reduction of the duty on monazite sand from 6 cents a pound to 4 cents a pound in 1909, the imports of thorium nitrate began to decrease, the requirements for this chemical being furnished by increasing imports of monazite sand. The imports of thorium nitrate decreased rapidly until in 1918 no imports were recorded.

During the next three years there was a decided growth in the import trade in this compound, but with the placing of monazite on the free list in 1922, they again decreased, while the imports of monazite sand increased.

Tables 11 and 12 show the imports of thorium nitrate, thorium oxide, and other thorium salts into the United States for the years 1901 to 1928, inclusive.

With the formation of the Thorium Cartel in 1902, the exports of thorium nitrate from the United States ceased.

In 1922, however, 1,103 pounds of thorium nitrate, valued at $3,149, were reported as being exported from the United States.

Table 11. - Thorium nitrate imported for consumption in the United States, 1901-1928[1]

Year	Quantity (pound)	Value	Price per pound
1901	[2] 12,986	$ 41,663	- -
1902	42,815	131,350	$ 4.00
1903	64,520	232,155	5.86
1904	58,655	249,904	5.98
1905	52,378	269,504	6.53
1906	40,090	139,929	3.78
1907	51,441	152,666	4.29
1908	65,289	173,239	4.70
1909	127,833	236,057	3.87
1910	124,808	219,615	3.10
1911	121,111	238,841	3.00
1912	117,485	225,386	3.00
1913	119,044	230,628	2.73
1914	101,927	239,376	2.98
1915	67,247	170,268	4.50
1916	909	3,884	6.25
1917	1,188	2,359	8.00
1918	- - -	- - -	8.00
1919	3,307	5,250	3.75
1920	25,640	43,221	1.21
1921	44,554	89,672	2.01
1922	43,254	42,096	.97
1923	24,027	29,789	1.24
1924	14,150	13,721	.97
1925	21,519	22,661	1.05
1926	21,403	27,955	1.31
1927	10,492	13,514	1.29
1928	12,431	14,149	1.14

[1] Mineral Resources of the United States, Part I, Metals.

[2] Fiscal year.

Table 12. - Thorium oxide and other salts imported into the United States, 1909-1928.[1]

Year	Quantity (pounds)	Value
1909	17,549	$ 19,596
1910	5,234	8,500
1911	- - -	- - -
1912	- - -	- - -
1913	1,763	2,694
1914	763	1,687
1915	139	349
1916	4,099	9,487
1917	11	923
1918	28	124
1919	- - -	- - -
1920	- - -	- - -
1921	- - -	- - -
1922)	(7,830	12,329
1922)	[2](52	20
1923	8,901	9,745
1924	26	33
1925	1,230	1,348
1926	13	20
1927	302	377
1928	9	47

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[1] Mineral Resources of the United States, Part II, Nonmetals.

[2] First figure covers Jan. 1-Sept. 21, 1922; second covers period of Sept. 22-Dec. 31, 1922.

MESOTHORIUM

Mesothorium is a radioactive element formed by the spontaneous disintegration of thorium and therefore is always found in thorium minerals. It is obtained as a by-product in the manufacture of thorium nitrate. The physical and chemical properties of mesothorium are in many respects identical with those of radium, and if the two are mixed intimately it is impossible to separate them. Mesothorium can be substituted for radium in therapy, but practically all of the mesothorium isolated is used to replace radium in luminous paint used for watch and clock dials and similar purposes.

If it is intended to recover mesothorium from monazite, a small quantity of barium chloride is added during the sulphuric acid treatment of the ore, and the mesothorium is removed with the insoluble barium sulphate. It is recovered from this precipitate and purified by a series of fractional crystallizations as the chloride, bromide, or other salt.

Imports

Table 13 shows the imports of radium salts and radioactive substances into the United States for the years 1918 to 1928.

Table 13. - Imports of radium salts and radioactive substances, into the United States, 1918-1928[1]

Year	Quantity (grains)	Value
1918	[2]10	$ 274
1919	13	11,651
1920	- -	- - -
1921	1,716	5,299
1922	12	24,476
1923	134	493,068
1924	125	408,146
1925	[3]154	241
1926	- -	671
1927	- -	522
1928	[4]169	565,537

[1] Commerce and Navigation, Annual of Department of Commerce.
[2] Radium salts and radioactive substances.
[3] Radioactive substances only.
[4] General imports; radium salts only.

PROPERTIES OF CERIUM

Metallic cerium is a steel-gray, lustrous, malleable, and very ductile metal which is soft enough to cut with a knife. In dry air the metal is moderately stable, but becomes superficially tarnished with yellow dioxide. Cerium is very slightly attacked by cold water; a slow evolution of hydrogen occurs in boiling water, and the metal is tarnished black. It is readily dissolved by dilute acids, but concentrated nitric acid produces a brown deposit consisting mainly of dioxide.

Cerium has a specific gravity of 6.9, and a melting point of 640° C.

The fact that cerium when alloyed with iron emits sparks if scratched or rubbed with a file or steel edge shows it to be admirably adapted for use as a sparking agent in automatic cigar lighters, etc.

CERIUM NITRATE

Description and Uses

Cerium nitrate is a white crystalline salt which is very deliquescent and decomposes if heated to 200° C. If it is to be used in the manufacture of gas mantles it whould be of a purity comparable to that required for thorium nitrate.

Probably even now the principal use for cerium nitrate is in gas-mantle manufacture, though substantial quantities are employed for the production of cerium metal for use in pyrophoric alloys.

This metal is said also to be used, in minor quantities, however, in the laboratory for the reduction of columbium, tantalum, etc. The oxidizing action of cerium compounds has led to their use in photography, as oxidizing catalysts in organic preparations, and as an ingredient in "driers." There is a possible chance that cerium oxides may be used in coloring glass, and cerium salts actually have been employed as mordants in dyeing. In the tanning industry, cerium salts have been used to a limited extent. The hide decomposes the salts, fixing it in the form of the hydrated oxide; it partly reduces the ceric sulphate, or ceric ammonium nitrate, absorbing oxygen in the process, and yields a good quality of leather which has a yellow tint and resists the action of water.

In addition to cerous oxalate, other salts - e.g., hypophosphate and double ammonium citrate, tartrate, and lactate - have been utilized medicinally in the treatment of seasickness and nervous disorders.

The rapidly growing use of cerium in the manufacture of pyrophoric alloys, however, has not as yet forced the industry to utilize monazite direct for the recovery of metallic cerium; the waste by-product from the manufacture of gas mantles are sufficient to supply the demands, as the gas-mantle industry utilizes only 1 to 2 per cent of the cerium in monazite.

PYROPHORIC ALLOYS

Description and Uses

In his research on the rare-earth metals, Welsbach discovered in 1903 that certain of the alloys when scratched with a file gave off sparks capable of igniti inflammable gas. Later discoveries and improvements demonstrated that an alloy of cerium metals with iron possesses the maximum pyrophoric properties.

This alloy, known as sparking alloy, misch metal, ferrocerium, or pyrophoric alloy, is used in automatic gas-lighting devices, miners' safety lamps, and cigar and cigarette lighters. It was used to a limited extent during the war in star shells, tracer bullets (the friction with the air causing it to flame brightl and in automatic lighters for use in trenches and other places where matches proved impracticable.

Method of Manufacture

The pyrophoric alloys may be made either from metallic cerium or from a mixture of the cerium-earth metals in approximately the following proportions: Cerium, 50 to 75 per cent; lanthanum, neodymium, and praesodymium, 25 to 45 per cent; and iron, 0.5 to 1 per cent.[11]

The manufacture of pyrophoric alloys employs the rare-earth residues obtained as a by-product from monazite sand, and first converts them to the anhydrous chlorides. The complete dehydration, which is a rather difficult process, is essential in the preparation of the material for the subsequent steps in its manufacture. The first step is to prepare the material for electrolysis; and it has been found that to do this suitably for the continuous commercial production of metallic cerium, certain special precautions are necessary. Certain rather limited ranges of temperature are required for efficiently performing the two processes carried out in the electrolytic cell, and these are called, respectively, "separating temperature" and "agglomerating temperature." The electrolyte is prepared preferably by using a solution of a mixture of chlorides of cerium and other rare-earth metals of which cerium is the chief constituent. It should be comparatively free from sulphuric acid and sulphate.[12]

The purity of this solution is not important, except for the percentages of sulphur and phosphorus compounds on the one hand, and for certain bases such as iron and aluminum compounds on the other, which should each be reduced below 3 per cent. An excess of cerium oxide will precipitate the iron and aluminum, and calcium chloride (or better, barium chloride) will throw down the sulphates and phosphates.

The solution is then clarified by filtering hot or settling, and evaporated to dryness. The preparation of the electrolyte should be so carried out as to secure the proper surface-tension conditions between the fused electrolyte and the fused metal when produced in the electrolytic bath. An excess of certain impurities, including oxychloride, tends to reduce the surface tension between the metal and electrolyte in the bath, and to alter the viscosity, producing an emulsion or colloidal solution of metal in the electrolyte, and preventing the separation of the metal from the bath.

The electrolysis is carried out in pots of cast iron high in carbon and silicon, about 8 inches in diameter and 12 to 18 inches in depth, usually set in brick work. The heat is applied externally and almost wholly at the bottom of the pot. A small amount of the electrolyte is first put into the pot and nearly fused by a gas heater. The electric current is then turned on and the fusion is completed. More electrolyte is gradually added, being brought to the fusion point each time, until the pot is full. Either carbon or graphite anodes are used.

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[11] United States Tariff Commission, Incandescent Gas Mantle Industry (Par. 154, Oct., 1913, Survey C. 22). Tariff Information Series, 1921, p. 27.

[12] Hirsch, Alcan, The American Pyrophoric-Alloy Industry. Chem. and Met. Eng., vol. 19, No. 62, Sept. 28, 1918, pp. 510-511.

The mixture of metals when it comes from the pot is relatively soft and does not spark easily upon scratching. To make the commercial product, therefore, it is alloyed with about 30 per cent of other metals, chiefly iron, and formed into small pieces to make the "flints" used in lighters, etc. The alloy enters into commerce in the form of small sticks or rods, either rectangular or round, and of various lengths, varying from 200 to 2,000 pieces per pound. Probably the commonest form is a round piece approximately one-eighth inch in diameter and one-eighth inch long; it takes from 1,500 to 2,000 pieces of this size to weigh 1 pound.

The manufacture of these small pieces from "misch metal" is a difficult operation, requiring delicate and expensive equipment.

Imports

Tables 14 and 15 show the imports of cerium metal and various cerium salts for the years 1922 to 1928, inclusive. These are principally from Germany.

Statistics are not available as to the domestic consumption of cerium and ferrocerium, but it is probably not in excess of a few thousand pounds annually.

Table 14. - Imports of cerium metal and of ferrocerium and other cerium alloys, 1922-1928[1]

Calendar year	Rate of duty	Quantity, pounds	Value	Average price per pound
Cerium metal:				
1922 (Sept. 22-Dec. 31)	$2 per pound	- - -	- - -	- -
1923	do.	38	$1,031	$27.15
1924	do.	- - -	- - -	- -
1925	do.	4	16	4.00
1926	do.	20	196	9.80
1927	do.	5	35	7.00
1928	do.	16	57	3.56
Ferrocerium and other alloys:-				
1922 (Sept. 22-Dec. 31)	$2 per pound and 25 per cent	- - -	- - -	- -
1923	do.	- - -	- - -	- -
1924	do.	100	208	2.08
1925	do.	89	185	2.08
1926	do.	1	22	22.00
1927	do.	71	118	1.66
1928	do.	2,085	5,999	2.88

[1] U. S. Tariff Commission, Summary of Tariff Information Schedule, No. 3, 1929, p. 624.

Table 15. - Imports of cerium nitrate, cerium fluoride, and other cerium salts, 1922-1928[1]

	Quantity (pounds)	Value
Cerium nitrate:		
1923	1,007	$ 866
1924	2,258	2,410
1925	5,124	5,535
1926	124	181
1927	20	34
1928	292	121
Cerium fluoride:		
1922 (Sept. 22-Dec. 31)	54	25
1923	38,971	3,058
1924	36,650	1,173
1925	- - -	- - -
1926	- - -	- - -
1927	39,683	2,469
1928	100	26
Other cerium salts:[2]		
1922 (Sept. 22-Dec. 31)	3	24
1923	4,821	746
1924	83,581	4,361
1925	16,969	1,487
1926	11,503	894
1927	47,155	3,627
1928	100,624	8,047

[1] U. S. Tariff Commission, Summary of Tariff Information Schedule, No. 1, 1929, p. 410.

[2] Not separately provided for.

PURCHASERS OF THORIUM AND CERIUM COMPOUNDS

Following is an incomplete list of possible purchasers of thorium and cerium compounds in the United States:

POSSIBLE PURCHASERS OF THORIUM AND CERIUM COMPOUNDS

Varlacoid Chemical Co.,
15 Moore St., New York, N. Y.

Gibson-Homans Western Co.,
3128 Roanoke Road, Kansas City, Mo.

Rex Research Corporation,
Toledo, Ohio.

A. Daigger & Co.,
159 West Kinzie St., Chicago, Ill.

A. D. Mackay,
26 Cortlandt St., New York, N. Y.

The Harshaw Chemical Co.,
150 Nassau St., New York, N. Y.

Foote Mineral Co.,
1608 Summer St., Philadelphia, Pa.

Mallinckrodt Chemical Works,
3600 N. 2nd St., St. Louis, Mo.

Maywood Chemical Works,
Maywood, N. J.

Merck & Company,
64 Park Place, New York, N. Y.

Pfaltz & Bauer, Inc.,
305 Pearl St., New York, N. Y.

Lindsay Light Co.,
161 E. Grand St., Chicago, Ill.

J. T. Baker Chemical Co.,
15 N. Broad St., Phillipsburg, N. J.

Harrison Manufacturing Co.,
Rahway, N. J.

Jungmann & Co., Inc.,
3 Desbrosses St., New York, N. Y.

Welsbach Co.,
Gloucester, N. J.

New Process Metals Corporation,
46-50 Center St., Newark, N. J.

FLAMING-ARC CARBONS AND GLOWER LAMPS

The flaming-arc electric lamp is based on the principal of introducing oxides of rare-earth metals into the carbons to secure an increased flame arc, somewhat as the Welsbach gas mantles increase the luminosity of a gas flame.

Calcium chloride and the oxides of iron, titanium, chromium, etc., were first employed with more or less success. In the making of the carbons (which are commonly made of petroleum coke, ground fine and mixed with tar, forced through dies and dried), the metallic oxides are either mixed with the mass or introduced as a core into the center of the carbon. An increased voltage is required, as the electrodes are drawn farther apart to get a long flame. In some forms, as the magnetite lamp, the lower (positive) electrode is simply a permanent knob of copper, while the upper (negative) electrode carries the mineral salts. Such lamps give a large body of arc or flame and do not burn a crater in the carbon but consume a comparatively flat surface when directly opposed. In one form two carbons are employed, positioned like the sides of a V. The quantity of vapor given off by the flame arc lamps necessitated an open globe at first, but later it was found possible to draw out the vapor by a draft and enclose the carbons in a suitable glass globe.

Another form of illumination using the rare-earth metals was the Nernst lamp. Instead of carbons it employed what is known as a "glower," this being a little rod, something over an inch long, made principally of the oxide of zirconium with varying amounts of rare-earth oxides, such as yttria, erbia, thoria, and ceria, mixed with a binding paste and baked into a material resembling porcelain. This rod was provided with platinum terminals. When a current was passed through it, it emitted a soft white light. The life of this form of lamp was from 400 to 600 hours - thus outlasting the arc-lamp carbons which have a life of 80 to 175 hours before they have to be replaced.

Prior to the growth of the pyrophoric-alloy industry the carbon lamp manufacturers were unable to absorb the entire waste by-product (the rare-earth metals) from the manufacture of gas mantles. The improved tungsten-filament electric lamp superseded the carbon arc lamp, and the by-product is now absorbed by the pyrophoric-alloy industry.

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Thorium

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